OPTICAL EXPERIMENTS WITH LASER SOURCES

Semiannual Technical Report

Covering Period Ending 28 February 1971

Due: 30 March 1971

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Sponsored by: Advanced Research Projects Agency
ARPA Order No. 675, Am. 4
Program Code Number 8EZ0

Contract No: DAHC04-69-C-0003

Effective Date of Contract: 20 August 1970 (continuation)

Contract Expiration Date: 20 August 1971

[redacted] [redacted]

Research Monitored by: U.S. Army Research Office - Durham
Box CM, Duke Station
Durham, North Carolina 27706

Report No. AROD 7871.4

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DAHC04-69-C-0003, U.S. Army Research Office

Walter L. Faust

Introduction

This semiannual report presents a discussion of technical findings and accomplishments for the period 30 September 1970 through 28 February 1971.

I. CO_2 Laser Work

A. Laser Development

By the present date, CO_2 lasers h een built in many laboratories, with widely varying designs, depending , on aspects of performance considered important n different applications. For the systems which we have developed, the prime consideration has bee.. wide spectral coverage in single line operation. Consistent with that limitation, we have sought maximal power output, for long-pulse and for Q-switch operation (thus, two distinct lasers have been built). It is likely that our lasers operate in the T.E.M $_{00q}$ modes, but no beat experiments have been performed to verify this. Stability and sealed-off operation have not been sought. The 4.5 meter resonator length which we employ for our purposes pro ably does not lend itself to extremely stable operation for two reasons: i) Mechanical rigidity is more difficult to achieve in such a length, and ii) The high gain and small C/2L encourage operation in more than one longitudinal mode. There is no apparent incompatibility between our techniques and sealed-off operation.

In addition to long discharge path, our lasers are characterized by

i) Minimal optical losses consistent with grating operation (for the Q-switched laser this involved some innovation), ii) A discharge operating in 300-millisecond pulses, and iii) Cooling of the gas discharge. All these encourage good operation even on weak lines, such as those near band-center (small J) or of large J.

For the long-pulse laser (which we call NQS for non-Q-switched), our techniques have led to operation on the record number of 130 lines. On each of the vibrational transitions 00°1 → 10°0 and 00°1 → 02°0, this laser misses no lines around band center; it operated in each case from R(0) to R(64) and from P(2) to P(66). Altogether we have dense coverage from 9 μ to 11 μ, at typically 1.5 cm^{-1} intervals, and with 10- to 20-watt bursts of 300-millisecond duration.

The Q-switched laser (QS) probably holds the record number of lines for its class of operation, also, but it is more difficult to search out prior claims in this case. We have 80 lines operating as follows: P(4) to P(42) and R(4) to R(42) for each of the two vibrational transitions. The lesser number of lines (80 < 130) for this laser is attributed to the problem of small gain and limited time for build-up from noise. This effect was discussed by Meyerhofer,[1] though not in the present context of limiting the number of lines. We verified this by "priming" introducing a small amount of radiation from NQS into QS, both adjusted for the same line. Indeed, we found i) fifteen lines which gave no observable energy previously, now give Q-switched pulses, ii) sixteen lines which ran weakly now give substantially larger pulses, and iii) strong lines experience no enhancement.

The above and other information on the two lasers was published in two papers during 1970.[2,3] The performance considerations given for the lasers were dictated by our intention to perform a resonance experiment on the simple vibrator system $CaF_2:H^-$. This experiment occupied us during the fall of 1970 and led to the results and publication discussed below in Part B.

For the purposes of a new sort of experiment, which we intend to describe in a later proposal, we have now a need to extend the NQS laser to continuous operation, retaining single-line sepctral-coverage and maximal power. These have been our design objectives during a period of moving to a new laboratory and rebuilding, since January. The new NQS laser will incorporate a water-cooled diffraction grating generated in metal on a copper substrate. It will have improved cooling of the gas discharge. And the electric discharge supply, which need not now be pulsed but which must handle a heavier average load, must have a new ballast system.

Toward these same design considerations of single-line spectral-coverage and high-power, we have been building since June 1970 a transverse-discharge, transverse gas-flow laser. Though it may be operated as a TEA laser, it is not limited to that; and we intend to try continuous operation at lower pressures. This machine, like NQS, is to be used with the copper grating discussed above. Despite some indications to the contrary,[4] we feel that such a system may have rather high specific gain. Thus, it may run well even on fairly weak lines and again give good spectral coverage. The main structure is complete, but work on the gas plumbing and electrical supply

remains. The most expensive item, a Rootes blower, was obtained at zero cost from surplus. It is in operation presently, driven by a 7.5 hp motor and V-belt. Because of the scanty information given in previous publications on similar lasers, there are several points of uncertainty in our design.

B. CaF_2H^- Experiment

This work has recently appeared in a publication entitled "High Intensity Resonance on the Simple Vibrator $CaF_2:H^-$".[4] The system affords a "textbook" problem - a spherical harmonic oscillator, slightly perturbed. Since the various $\Delta n = 1$ transitions have slightly different frequencies, they can be distinguished from one another. In principle they might also be distinguished by differing polarizations, but we will see that a "scrambling" process prevents this in fact. Several studies we performed are:

i) Rough measurements of the saturation parameter W which enters the relation for absorption by defect centers

$$(1) \qquad (1/I)\,(dI/dx) = -\alpha_H/(1 + W)\ ,$$

for the $n = 0$ to $n = 1$ frequency, ω_{01}.

For large I such that $WI \gg I$, we have $-dI/dx = \alpha_H/W$. The left side is clearly just the absorbed power per unit volume. In this same high-intensity limit we can see also that $-dI/dx = (1/2n)\hbar\,\omega_{01}/T_1$. Here n is the volume density of defects. In saturation, one-half of them are excited at any moment. Each contributes a power absorption given by $\hbar\omega_{01}$ divided

by the vibrational decay lifetime T_1. This limiting behavior is presented also in more detailed theories for a homogeneous resonance. Using then the definition of the absorption cross section $\sigma \equiv \alpha_H/n$, we find an expression for the decay time T_1 of a center, independent of n (saturation is a private activity of each center individually).

$$(2) \qquad T_1 = \frac{1}{2} \hbar\, \omega_{01}\, W/\sigma \quad .$$

The cross section was calculated from data of prior workers.[5] W was determined by study of the dependence of the fractional transmitted intensity upon the incident intensity, using essentially (but not exactly) the integral of Equation (1). But the uncertainty is large, perhaps a factor of three. This is the case because it is difficult to assign a spot diameter d for the focussed beam. Thus, though our measurements of total power are fairly reliable, the focussed intensity is difficult to estimate, and the uncertainty of course propagates to W. The difficulty has previously appeared in other contexts, and it is not insurmountable. For instance, in an earlier experiment with mixing of 6328 Å and 28μ light, we obtained photographs of the infrared spot.[6] We have not undertaken such elaborate measurements for the present purposes. We have simply taken $d = 80\mu$, by "dead reckoning" on prior experience. We are content with the consequent rough estimate from Equation (2): $T_1 \simeq 7$ psec.

The uncertainty principle then dictates a minimal line width of $(2\, C\, T_1)^{-1}$ in wave number units; that is, 0.7 cm^{-1}. This is in fact just the observed line width in the limit of low temperature. The entire

picture is in accord with the ascertion by Hayes, et al.,[7] that the residual width is due to decay. There is no inhomogeneous broadening as from strain. Because of the mode of preparation and the nature of the defect, the environments of all centers are expected to be quite identical. Such a decay line width obviously would be homogeneous. We have in fact several indications that the line is homogeneous, at least on any time scale significant in our experiments. For instance, Equation (1) would not describe saturation for an inhomogeneous line. Hayes, et al., continue to state that the "temperature-dependent [excess] ... width" is due to phonons scattered [~ elastically] without change in the vibrational state. But, with randomization of phase, corresponding to the excess of $1/T_2$ beyond $1/T_1$, we might add.

ii) Our center has an additional sort of relaxation beyond that for the usually discussed non-degenerate two-level system described by T_1, for energy loss, and T_2 (necessarily $\leq T_1$) for phase coherence loss. Because of the x, y, z degeneracy for the n = 1 excitation, this center is able also to forget the polarization of some initial excitation. Briefly, if we excite an n = 1 excitation in a specific polarization, there should be selection rules for a subsequent n = 1 to n = 2 excitation with polarized light. But these all are violated, indicating scrambling in the n = 1 level. It is our belief that the same elastic phonon scattering processes account for this effect as for the phase coherence loss, so that the same relation time T_2 should describe it.

iii) In our publication[4] we continue to discuss several other features of the system, such as frequency additivity: $\omega_{01} + \omega_{12} = \omega_{02}$.

That this is satisfied is a consequence of the "no-phonon" character of the resonances we are driving.

II. Light Scattering from Semiconductors, Associated with Impurities

In the previous report we described studies of Raman scattering from localized vibrational modes associated with impurities in GaP. This work appeared in print[8] shortly after the previous Semiannual Technical Report. The technique may be of serious utility in assay of impurities in semiconductors, commercially. We have described several advantageous features of the technique: i) Small sample volumes may be probed, ii) One can work with material of a free carrier concentration so large that infrared transmission would not be possible, iii) One can obtain information about concentrations of impurities on specific sites, once the resonances have been identified (it also does this), iv) From study of the polarization of the scattered light one can make choices amont possible sites for the impurity. As of the previous report, we had observed first high-frequency vibrations (above all phonons) and then vibrations of frequencies in the gap between the acoustical and the optical phonons - in each case in GaP. We have now successfully extended the method to the study of impurities in GaAs, where one must use infrared light; the wavelength must extend that of the band gap, ~8400 Å for the light to be transmitted. For this we used a borrowed Nd laser. Only high frequency modes have thus far been observed. This extension to GaAs may be important because this material has considerable interest for the microelectronics industry.

We have pursued studies of free carrier plasma and lattice vibration interaction in GaP, paralleling in some degree those of Mooradian for GaAs. It happens that the spectral features observed with GaP have strong qualitative differences from those of GaAs. We will outline briefly a simple mathematical dielectric theory which accounts for the spectra of both sorts in a unified way. In general, the complex dielectric susceptibility may be displayed as

$$\varepsilon(\omega) = \underbrace{\varepsilon_\infty \left(1 + \frac{\Omega^2}{\omega_T^2 - \omega^2 - i\omega/\tau_L}\right)}_{\text{lattice vibration}} - \underbrace{\frac{\omega_p^2}{\omega^2 + i\omega/\tau_p}}_{\text{free carriers}} .$$

Resonances are associated with zeros at $R_e\ \varepsilon(\omega)$.

$$\text{Re}\ \varepsilon(\omega) = \underbrace{\varepsilon_\infty \left(1 + \frac{\Omega^2(\omega_T^2 - \omega^2)}{(\omega_T^2 - \omega^2) + (\omega/\omega_L)^2}\right)}_{\text{lattice}} - \underbrace{\frac{\omega_p^2}{\omega^2 + 1/\tau_p^2}}_{\text{free carriers}}$$

We consider frequencies $\omega < \omega_T$. The lattice contribution is necessarily positive. One may appreciate that, for the case of large τ_p, there may exist a sufficiently small ω that the (negative) free carrier term becomes just large enough to cancel the lattice term. This seems to be the case for GaAs. In GaP, τ_p is known to be small; and one may appreciate that in this case there may exist no ω which can give a large enough free carrier term to produce cancellation; the $1/\tau_p^2$ contribution to the denominator is too

large. In fact, no plasma resonance is observed with GaP. The cases of large and of small τ_p are presented respectively in Figures 1(a) and 1(b). Not only is there no plasma resonance for GaP, but one may see also that the apparent LO frequency experiences, in this case, only a relatively small displacement in virtue of interaction with the plasma. We are completing Hall and conductivity measurements to demonstrate quantitative agreement with the theory; preliminary results are good.

III. Far Infrared Work

We have obtained operation at the relatively low level of ~100 watts from our large H_2O laser. We have also in operation a pulsed argon laser which generates ~40 watts at 4880 Å and 5145 Å. We intend to use these two together, to study phonon sidebands in excition fluorescence excited by the argon laser.

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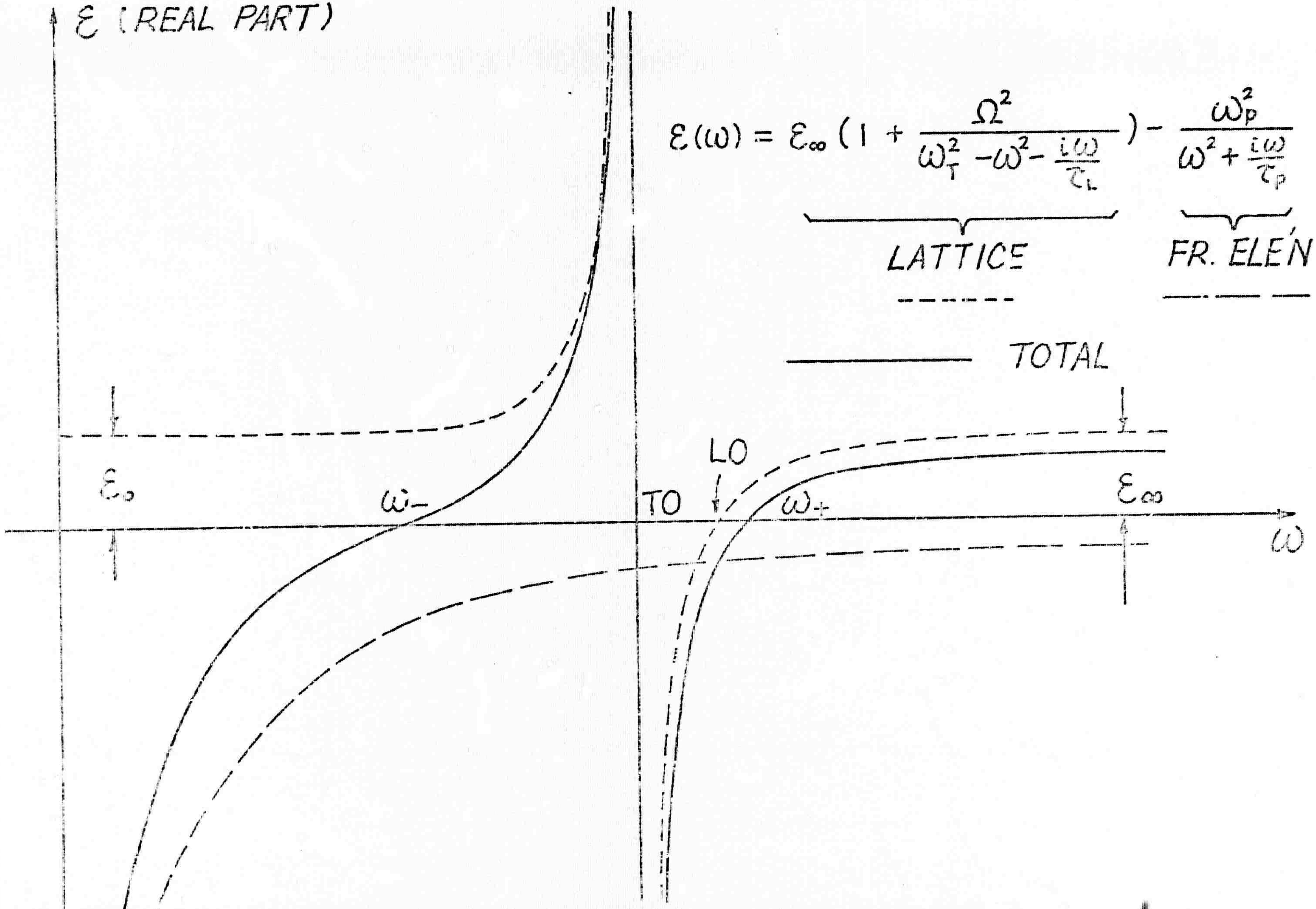
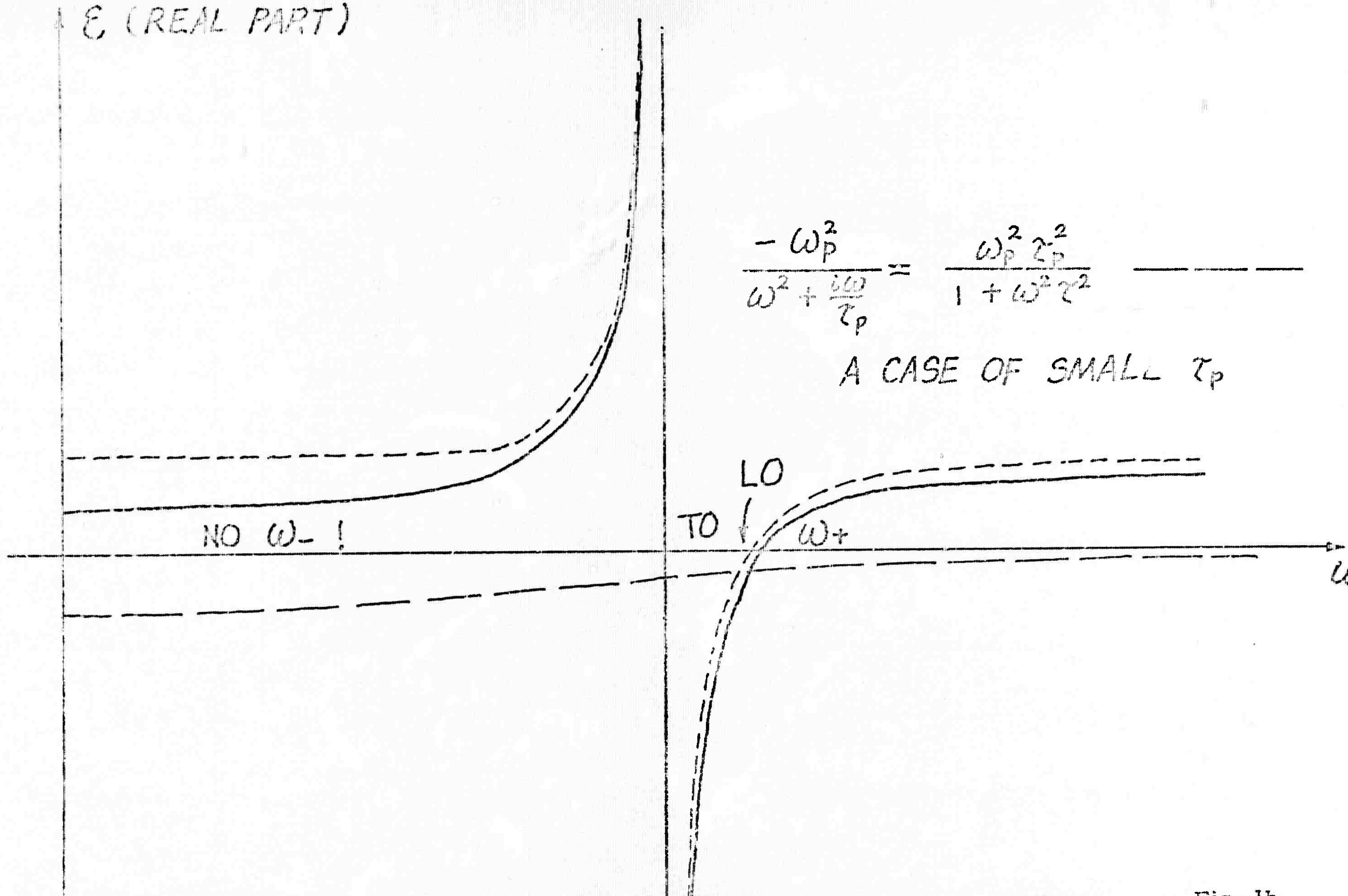

ε (REAL PART)
$\varepsilon(\omega) = \varepsilon_\infty \left(1 + \frac{\Omega^2}{\omega_T^2 - \omega^2 - \frac{i\omega}{\tau_L}}\right) - \frac{\omega_p^2}{\omega^2 + \frac{i\omega}{\tau_p}}$
LATTICE
FR. ELEN
TOTAL
ε_0
ω_-
TO
LO
ω_+
ε_∞
ω

Fig. 1a



ε (REAL PART)
$\frac{-\omega_p^2}{\omega^2 + \frac{i\omega}{\tau_p}} = \frac{\omega_p^2 \tau_p^2}{1 + \omega^2 \tau^2}$
A CASE OF SMALL τ_p
LO
TO
ω_+
NO ω_- !
ω

Fig. 1b